AEC RESEARCH AND DEVELOPMENT REPORT

REMOVAL OF LOW-LEVEL RADIOACTIVE WASTES BY A SANITARY WATER TREATMENT PROCESS

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A B S T R A C T

This report presents a study of the "cleanup factors" which were obtained across a potable water treatment plant using normal treatment methods; the average gross beta radioactivity was reduced by 15% and the strontium beta concentrations by 6%. A summary and evaluation of six months data obtained from daily field samples of the raw water supply, as well as the finished water product, is included. Sample analysis included determinations for gross beta radioactivity and radiostrontium. The raw water radioactivity concentrations were less than 25% of the maximum permissible concentrations in water recommended by the National Committee on Radiation Protection.

REMOVAL OF LOW-LEVEL RADIOACTIVE WASTES BY A SANITARY WATER TREATMENT PROCESS

INTRODUCTION AND SUMMARY

The effects of various applications of coagulation, softening, and precipitation in water treatment processes upon radioactive liquids have been previously described by the U. S. Public Health Service and the U. S. Army, as well as several AEC facilities.[1,2,3,4,5] The removals achieved varied from 0% to 99% for the isotopes studied. For the most part, these removal efficiencies have been ascertained on a laboratory scale. In a few cases, pilot-plant units have been used to extend the study of coagulation, filtration, and softening processes using variable chemical dosages, coagulants, pH conditions, and radioactivity concentrations. For this study, the results were obtained from a full-scale sanitary water treatment plant in routine daily operation to provide potable water for an industrial plant with a population of about 5,000 employees. The treatment facility is located on the Clinch River about 6 miles downstream from another nuclear facility where low-level radioactive wastes are discharged to the stream at levels well below the maximum permissible concentrations in water (MPC_w) recommended by the National Committee on Radiation Protection (NCRP) for the mixture of radionuclides of concern.[6] The gross beta radioactivity concentration (primarily ruthenium and strontium), which included that of the river background due to naturally occurring radioelements and fallout from nuclear weapons tests, represented about 15% of the appropriate MPC_w. Under these conditions, the "cleanup factor" for this sanitary water treatment plant under normal operating conditions over a 6-month period averaged 15% for the gross beta activity, with a comparable value of 6% for the $^{89-90}Sr$ beta activity.

DISCUSSION

Nowadays, we take as a matter of fact the purity of the water we drink. We assume, and rightly so, that our health is being protected by the carefully guarded provision of a clean, pure water supply. Even so, nearly all water sources contain small, but measurable, amounts of radioactive material. As rain seeps through the soil, it dissolves many types of minerals, some of which are radioactive, including uranium, thorium, and potassium. Raindrops condensing on air-borne radioactive fallout bring particles to the earth, including strontium and cesium, which eventually find their way into streams and ponds. In most cases, this background radiation is insignificant and no health hazards are indicated. Radioactivity levels in the environment, while detectable, are variable and relatively low, as reflected in Figure 1, Beta Radioactivity Levels in Streams and Periods of Nuclear Tests.[7]

Stream survey is a key means of delineating a specific waste disposal problem, as well as determining the extent and effects of radioactive

contamination on pollution. With the advent of nuclear energy for both military and civil uses, measurements of water quality in streams and waterways will of necessity have to include the various radionuclides of interest, thus requiring modification of present water works procedures and the development of simplified detection and control systems for such materials.

Water pollution control agencies are understandably concerned with radioactive liquid waste discharges that may interfere with downstream water uses. (The attention and "concern" are frequently not in proportion to the potential problem, probably due to the romance and mystery of the atom bomb and the association of all ratioactivity with nuclear weapons and explosions). On the other hand, we must also learn how best to protect our domestic water supply, as well as water used for agriculture, industrial processes, recreational purposes, and the support of aquatic life.

The effects of water treatment processes on radioactive liquids have been studied by several agencies and scientific groups. Different processes have been found to remove various amounts of radioactivity. Except for ion-exchange methods, they do not remove sufficiently significant amounts of the critical isotopes, such as cesium and strontium, to justify reliance upon them as a protective factor. For the most part, these removal efficiencies have been ascertained on a laboratory scale. In some cases, pilot-plant units have been used to extend this study on coagulation, filtration, and softening processes. In our case, we have studied the effects of the routine day-by-day operation of our sanitary water treatment plant on the concentrations of the radionuclides in the influent stream. The raw water intake and the corresponding finished water were analyzed for gross beta radioactivity, as well as the $^{89\text{-}90}Sr$ content, thus providing an evaluation of the effectiveness of normal sanitary water treatment for the removal of radionuclides from water.

WATER TREATMENT PLANT

The source of our raw water is the Clinch River at a point 65 miles below its impoundment by the Norris Reservoir and 6 miles downstream from another nuclear facility where low-level radioactive wastes are discharged to the stream at levels well below the MPC_w recommended by the International Commission on Radiological Protection (ICRP) and the NCRP for the mixture of radionuclides of concern. (See Figure 2, Map - Lower Clinch River Basin). Backwaters formed by Norris Dam extend 70 miles in the Kentucky-Tennessee area, and between the impounded lake and the head waters of the Clinch River, which rises near Tazewell, Virginia, there are no metropolitan areas of a significant size, and there are thus no known sources of radioelements being discharged into the stream outside of the Oak Ridge area. This source of raw water is relatively pure and lacking in the usual industrial wastes and sewage which are so often found below large cities on some of our major surface waterways.

The sanitary water system, including the filtration plant, is operated by the Utilities Department of the Production Division at the Oak Ridge Gaseous Diffusion Plant. (See Figure 3, ORGDP Potable Water Treatment Plant). Two 3,200-gallon-per-minute deepwell turbine pumps at the raw-water pumping station deliver the water to a 150,000-gallon raw-water aeration and presedimentation tank. From here, the raw water flows by gravity into the filtration plant where coagulating and sterilizing chemicals are added. The coagulant is filter alum (aluminum sulfate) and the sterilizing agent is chlorine. The necessary amounts of these chemicals to be added varies with the turbidity, alkalization, and chlorine demand of the raw water. These amounts are determined by chemical tests performed by the filter plant operators. After addition of the necessary chemicals, the water enters the coagulation basins where a gelatinous floc of aluminum hydroxide is precipitated and adsorbs and encloses the suspended impurities, including bacteria. Slowly turning paddles bring this floc into intimate contact with the water, allowing the floc particles to become larger and heavier.

Following the coagulation basins, the water flows into the sedimentation basins where, in the absence of moving paddles, all of the undissolved material is allowed to settle. The clear water leaves the sedimentation basins and then flows to the top of the filter beds inside the filtration plant. The filtering facilities consist of five sand beds through which the water filters and flows to one of the two clear wells. For removal of residual bacteria, the clear water is treated with liquid chlorine and the potable water is transferred by pump to an elevated $1\frac{1}{2}$-million-gallon concrete storage tank for distribution through the plant.

WATER SAMPLING

In addition to a commercially procured, continuously recording and alarming beta water monitor, routine environmental monitoring of the radioactivity in the raw water supply (Clinch River) and the finished water at the treatment plant consists of weekly laboratory analysis of a composite of the continuous sample obtained during the previous seven days. In addition, during the 6-month period from October 18, 1962, through April 19, 1963, daily sampling of the beta radioactivity in the raw water and in the finished water was conducted to monitor the possible effects of TVA operations upstream in the Clinch River. These activities included the dredging of navigational channels, the construction of Melton Hill Dam, and the initial impoundment of water behind this dam. Associated with these activities were controlled fluctuations of Norris Dam discharge, which at times resulted in negligible Clinch River flow with resultant minimization of the dilution factor for the upstream radioactive waste discharge. Under these conditions, the downstream Watts Bar impoundment tends to flow upstream in the Clinch River, and the probable dispersal of wastes in the stream cannot be reasonably evaluated.

This opportunity was taken to (1) record both the gross beta activity and

the strontium beta activity levels in the raw water and in the finished water of the ORGDP Sanitary Water Treatment Plant, (2) evaluate the possibility of a correlation between the total beta activity and the corresponding strontium beta radioactivity present in the raw water supply, and (3) determine the beta radioactivity "cleanup factor" for this plant under normal treatment procedures. The water distribution system was also sampled periodically at dead-end sections of the duct runs, the cafeteria cooking water, and drinking water fountains.

MEASUREMENT OF RADIOACTIVITY AND RADIOCHEMICAL ANALYSES

The laboratory procedure for total beta counting of water samples involves evaporating to dryness a 100 ml. aliquot. The residue is dissolved in nitric acid and transferred to a stainless steel dish. This solution is then evaporated to dryness in the dish and the residue is beta counted in a proportional counter. At the time of this study, the precision of a single analysis at the 95% confidence level was ± 14% of the value, and there was no significant bias. Improved precision is currently being experienced.

The method for the determination of radiostrontium in water[8] in the presence of other radioelements at concentrations as low as 10 μμc/liter (22 beta disintegrations/minute/liter), at the drinking water levels prescribed by the U.S. Public Health Service, involves the addition of a known amount of natural strontium to the sample as a carrier. Yttrium-90, the daughter of ^{90}Sr, is removed by scavenging with iron. The strontium is then separated from impurities by a triple precipitation; first as the oxalate, then as the nitrate in concentrated nitric acid, and again as the oxalate which is dried, weighed, and beta counted. The activity of the recovered ^{90}Sr is determined from either an absorption curve technique or from the growth and decay curves of ^{90}Y and ^{90}Sr, respectively.

EVALUATION OF DATA

The data for this study indicate that:

1. The average strontium beta concentration in the Clinch River, by coincidence, approximated the 10 μμc/liter (10^{-8} μc/cc) value suggested by the U.S. Public Health Service for 168-hour week, non-occupational exposure to ^{90}Sr. (See Table 1). It should be noted that checks during this period showed that the ^{90}Sr contribution to the total strontium beta activity in the raw water supply varied between 25% and 40%, and averaged about 30%, indicating that the major portion of the strontium activity was from nuclear weapons testing involving ^{89}Sr from the fallout. For purposes of comparison, it is noted that the NCRP MPC_w for the 168-hour week general population exposure for ^{90}Sr is 4×10^{-7} μc/cc and the MPC_w for ^{89}Sr is 10^{-5} μc/cc; the former obviously is the controlling health factor.

During this 6-month period, the beta radioactivity represented about 15% of the NCRP MPC_W for the mixture of radionuclides in the Clinch River. The beta radioactivity data for the ORGDP potable water are shown in Table 2.

TABLE 1

BETA RADIOACTIVITY IN THE CLINCH RIVER
(October 1962 - April 1963)

	Monthly Average							Daily Sample		
	Oct	Nov	Dec	Jan	Feb	Mar	Apr	Max.	Avg.	Min.
Gross Beta* (10^{-8} μc/cc)	6	72	50	43	54	25	41	260	44	4
Strontium Beta (10^{-8} μc/cc)	0.5	1.7	1.0	1.0	1.2	1.2	1.5	3.2	1.2	0.3
Ratio - Gross Beta to Strontium Beta	12	42	52	43	45	21	27	52	37	12

*MPC_W (Mixed beta emitters; general population) = 350 x 10^{-8} μc/cc.

TABLE 2

BETA RADIOACTIVITY IN THE ORGDP POTABLE WATER
(October 1962 - April 1963)

	Monthly Average							Daily Sample		
	Oct	Nov	Dec	Jan	Feb	Mar	Apr	Max.	Avg.	Min.
Gross Beta* (10^{-8} μc/cc)	6	56	41	42	49	16	33	270	37	2
Strontium Beta (10^{-8} μc/cc)	0.4	1.3	1.0	1.1	1.2	1.1	1.3	3.1	1.1	0.2
Ratio - Gross Beta to Strontium Beta	14	43	41	38	41	15	25	43	34	14

*MPC_W (Mixed beta emitters; occupational) = 3500 x 10^{-8} μc/cc.

2. The "cleanup factor" for the beta radioactivity in water by the ORGDP Sanitary Water Treatment Plant under normal operating conditions (see Table 3) varies from 0% to 36% and averages 15% in the case of total beta radioactivity. Similar reductions are reflected by 10-year data

of monthly averages (see Table 4). Comparable figures for strontium beta ranged from 0% to 24%, with an average of 6%. The pH of the Clinch River varied between 7 and 9, averaging 7.8 during this period. Turbidity averaged 28 ppm, with 15% of the solids suspended and 85% dissolved. In the event of significant contamination of the water supply by these materials, adjustment of the pH, plus addition of water softeners and other flocculating chemicals, would be expected to precipitate more of the calcium compounds and the absorbed strontium, thus providing somewhat more effective reductions in the total beta radioactivity in the finished water.

TABLE 3

REDUCTION OF BETA RADIOACTIVITY BY ORGDP POTABLE WATER TREATMENT PLANT (October 1962 - April 1963)

	Oct	Nov	Dec	Jan	Feb	Mar	Apr	Total
Number of Samples	18	29	29	30	26	28	16	176
Reduction of Gross Beta Activity	0%	22%	18%	2%	9%	36%	20%	15% ± 24% @ 95%
Reduction of Strontium Beta Activity	9%	24%	0%	0%	0%	8%	13%	6% ± 17% @ 95%

TABLE 4

REDUCTION OF TOTAL BETA RADIOACTIVITY IN THE ORGDP POTABLE WATER TREATMENT PLANT FOR THE TEN-YEAR PERIOD, 1955-1964

Water Sampled	Ratio	"Cleanup" Factor
Raw/Finished	1.2 ± 0.6	13% ± 7% @ 95% C.L.
Finished/Potable	1.1 ± 0.7	6% ± 4% @ 95% C.L.
Raw/Potable	1.2 ± 0.6	18% ± 10% @ 95% C.L.

NOTE: Beta radioactivity average concentrations detected represent from 2% to 20% of the MPC_w for 168-hour week exposure to the general population, with a peak of about 50% MPC_w for a single weekly sampling period.

3. There was no statistically significant correlation between the total beta radioactivity and the corresponding strontium beta activity in the Clinch River during this period. Were such a correlation present, water treatment plants using this stream for their raw water supply could routinely monitor for total beta activity and confidently control on this basis, until such time that relatively high total beta levels suggested the advisability of performing the more costly and analytically demanding strontium separation and analysis.

4. Exposures of water treatment plant personnel to external penetrating radiation were not significantly different from normal background values measured in this geographic area.

5. The water distribution system, comprising about 27 miles of pipe, was also sampled at dead-end sections of the duct runs, in the cafeteria cooking water, and at drinking water fountains. There was no detectable $^{89\text{-}90}Sr$ in the scale and residue found in the supply duct system.

SUMMARY

Our present knowledge indicates that conventional potable water treatment processes are relatively inefficient for removal of radioelements from the low-level wastes which can be expected to be found in raw water supplies. Thus, strict control of waste discharge concentrations must be maintained to assure the purity of future water supplies. Assuming that these recommended discharge levels are maintained, it is unlikely that additional, specific treatment will be needed. Rather, our primary job will be that of continuing effective monitoring of the water supply to reassure outselves that pollution control measures are in fact adequate. On the other hand, under emergency conditions, it is anticipated that significant reduction can be made in the levels of these radioactive constituents by some of the currently available methods.

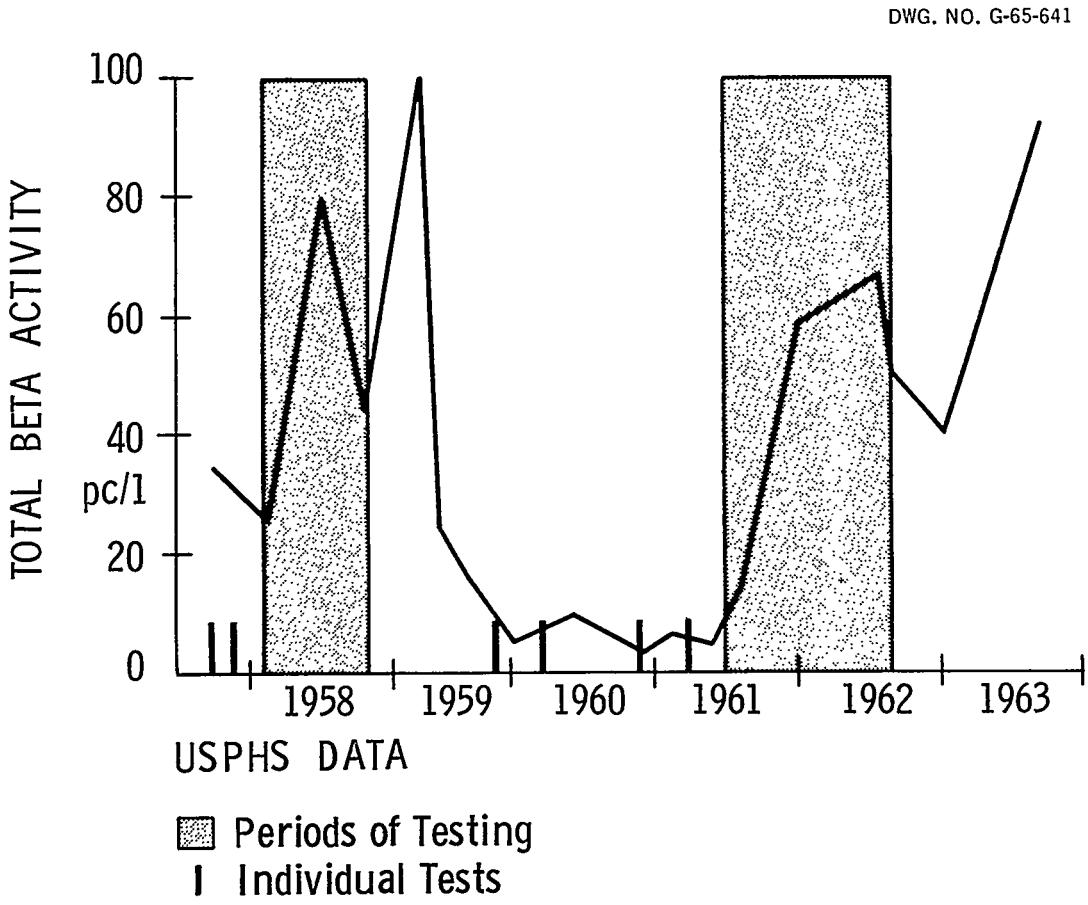

Figure 1

BETA RADIOACTIVITY LEVELS IN STREAMS
AND PERIODS OF NUCLEAR TESTS

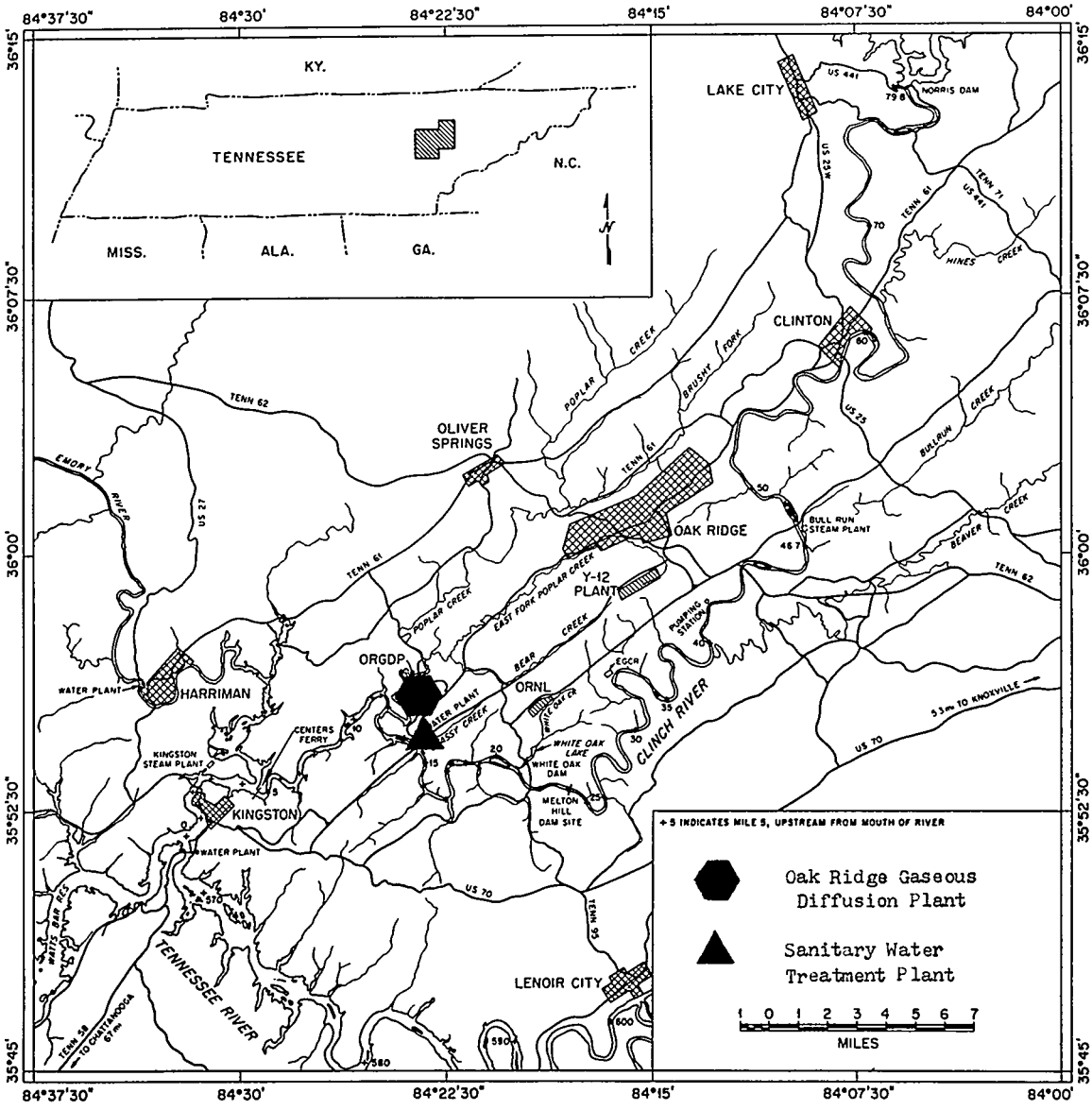

Figure 2

MAP - LOWER CLINCH RIVER BASIN

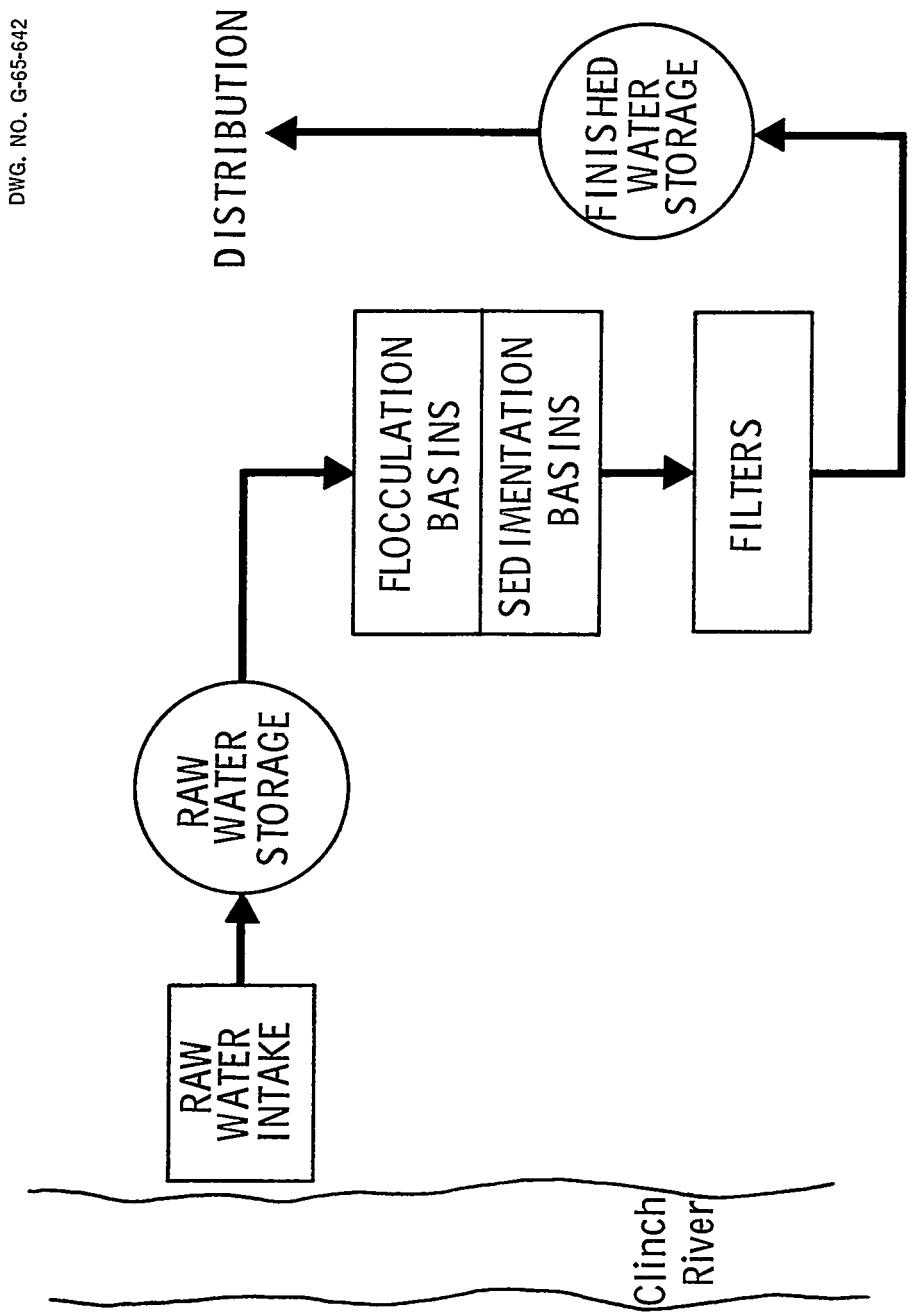

Figure 3
ORGDP POTABLE WATER TREATMENT PLANT

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